

REMARKS

This is a full and timely response to the outstanding non-final Office Action mailed November 2, 2005 (Paper No. 103005). Upon entry of this response, claims 46-50 are pending in the application. In this response, claim 46 and 50 have been amended. Applicants respectfully requests that the amendments being filed herewith be entered and request that there be reconsideration of all pending claims.

1. Rejection of Claims 48 and 49 under 35 U.S.C. §112, Second Paragraph

Claims 48 and 49 have been rejected under 35 U.S.C. §112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which Applicants regard as their invention. The Office Action states that:

Specifically, the language within lines 4 and 5 of claim 48 is unclear with respect to what is being reacted. The language “reacting a compound obtained by reacting a compound (eb) with a compound (fb) and further reacting with a compound (i)”, is confusing, because it cannot be determined what is being initially reacted with the “obtained” compound, since compound (i) is claimed as being “further” reacted. In other words, there appears to be too many occurrences of “reacting.”
(Office Action, p. 2)

Independent claim 48 is amended to further clarify the subject matter which Applicants regard as their invention. Applicants respectfully submit that the amendment overcomes the rejection of claims 48 and 49, and requests that the rejection be withdrawn.

2. Rejection of Claims 46 and 47 under 35 U.S.C. §103

Claims 46 and 47 have been rejected under §103(a) as allegedly obvious over *Barron* (U.S.4,067,844) or *Zwiener* (U.S. 5,364,955). Applicant respectfully traverses the rejection of claims 46 and 47. It is well established at law that, for a proper rejection of a claim under 35 U.S.C. §103 as being obvious based upon a combination of references, the cited combination of

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references must disclose, teach, or suggest, either implicitly, all elements/features/steps of the claim at issue. *See, e.g., In re Dow Chemical*, 5 U.S.P.Q.2d 1529, 1531 (Fed. Cir. 1988); *In re Keller*, 208 U.S.P.Q.2d 871, 881 (C.C.P.A. 1981).

a. Claim 46

The Office Action takes the position that “the respective processes [of *Barron* and *Zwiener*] yield the same product and the only difference amounts to changing the sequence of steps in a multi-step process, and it has been held that such a modification is obvious where an unexpected result is not obtained.” (Office Action, p. 3, section 4.) As will be explained in more detail below, Applicants respectfully disagree with this characterization of the references as yielding the same product as claims 46 and 47.

1) *Barron* and *Zwiener* Produce a Side Reaction

As argued in the last Office Action, both *Barron* and *Zwiener* disclose that a prepolymer is produced in step (2) of their respective processes. Applicants also argued that because a prepolymer is produced a side reaction such as dimerization and trimerization of urethane prepolymer inevitably occurs, so that a product produced by *Barron* or *Zwiener* has high molecule and high viscosity. (See Response filed Aug. 3, 2005, p. 8). In contrast, the process of claim 46 dispenses with the step of producing a prepolymer, and a product of claim 46 has low viscosity. Thus, the process of claim 46 differs in a non-obvious way from the processes disclosed in *Barron* and *Zwiener*.

The outstanding Office Action stated that Applicants’ arguments about side reactions were unsubstantiated opinion, since no evidence or logical rationale was provided to support Applicants’ position. (Office Action, p. 4.) Applicants provide herein both logical rationale and experimental data supporting Applicants’ position with regard to side reactions.

In a reaction process like *Barron* and *Zweiner*, when a polyisocyanate is reacted with a polyol, a prepolymer is produced. If this reaction progresses ideally, the NCO terminated prepolymer (basic unit) is obtained with reaction of two polyisocyanate molecules and one polyol molecule. However, as shown by experimental data in Appendix A-1, a dimer (double unit) and trimer (triple unit) which has twice or three times of molecular weight are also produced by side reactions, due to both polyisocyanate and polyol being multi-functional compounds. Accordingly, the final prepolymer product contains a large amount of high weight molecule.

This is evidenced by the experimental data presented in Appendices A and B. Appendix A is a Japanese language article from a 1996 issue of “Adhesion Technology Japan”, and Appendix B is the English translation of Table 3-18. Dimerization and trimerization in prepolymer production are clearly seen in the data of Appendix A/B, which will now be explained in detail. Table 3.18 shows the GPC (Gel Permeation Chromatography) charts of the prepolymer, with various isocyanate group contents which have been obtained using the same polyol (molecular weight 3000 of PPG) and polyisocyanate (polymeric MDI). In the Table, “iso monomer” means isocyanate monomer, “iso dimer” means dimer of isocyanate, “iso trimer” means trimer of isocyanate, “iso tetramer” means tetramer of isocyanate, “basic unit” means monomer or basic unit (prepolymer), “pre dimer” means dimer (double unit) of prepolymer, and “pre trimer” means trimer (triple unit) of prepolymer.

In the Table, chart (a) shows the prepolymer (15% isocyanate group content) which is obtained by 2:1 reaction with NCO group of polyisocyanate and OH group of polyol. Even in such prepolymer prepared at the theoretical reacting ratio, peak (7) of “pre dimer (double unit)” is observed. Further, as the isocyanate group content in the prepolymer is decreased from 15% to

6% the content of “pre dimer (double unit)” (7) and “pre trimer (triple unit)” (8) is increased. In any case where isocyanate group content is 15%, 9% or 6%, the dimer and/or trimer exist in the prepolymer product.

In other words, the side reaction, that is, production of dimerization and trimerization, *inevitably* occurs in producing a prepolymer by reacting a polyisocyanate with a polyol. Accordingly, an actual pre-polymer composition has higher molecule weight and higher viscosity than those of the ideal basic unit. Also, the final product or resin compositions, which use the prepared prepolymer as raw materials, have high viscosity. As is apparent from the above, the occurrence of side reactions is already known, and is supported by Appendix A/B.

The process of claim 46 therefore differs in a non-obvious way from the processes disclosed in *Barron* and *Zwiener*. Accordingly, a *prima facie* case establishing an obviousness rejection under *Ex parte Rubin* has not been made. For at least these reasons, claim 46 is not obvious over *Barron* or *Zwiener*, and the rejection should be withdrawn.

2) *Barron* and *Zwiener* Produce a Different Product than Claim 46

As argued in the last Office Action, according to the process disclosed in *Barron* and *Zwiener*, the urethane polymer has a hydrolysable silyl group and a NCO group in the terminal. On the other hand, the product of claim 46 has a hydrolysable silyl group and an OH group in the terminal. (See Response filed Aug. 3, 2005, pp. 8-9.) Thus, the products of *Barron* and *Zwiener* are different from the product of claim 46.

The outstanding Office Action stated that “applicants’ arguments are largely based upon the position that the instant products have an OH group in the terminal; however, applicants’ claims are not so limited. There is no requirement that such a terminal OH group exists or be produced.” (Office Action, p. 4.) Applicants provide herein a logical rationale supporting Applicants’ position.

Considering the process of the reaction, it is clear to one of ordinary skill in the art that the product by the process of claim 46 necessarily contains hydrolysable silyl group terminated polymer, and may in some circumstances contain OH group terminated polymer. However, applicants do not limit the product of claim 46 to the products having a terminal OH group. Applicants' reference to the "OH group" in the last response was made to emphasize the difference from *Barron* and *Zwiener*. The final product of claim 46 is hydrolysable silyl group terminated polymer, and if all ends of polyol do not react, the product may have a terminal OH group. That is simply an issue of possibility or probability.

Specifically, the process of Claim 46 comprises (1) obtaining a silicon compound (product (N)) having an active hydrogen being reactive with a isocyanate group, (2) reacting the product (N) with a compound (G) having an NCO group to obtain a silicon compound (product (O)) having a NCO group, and (3) reacting the product (O) with a compound having an OH group. The final product is hydrolysable silyl group terminated polymer. and if all ends of polyol do not react, the product may have a terminal OH group. See Appendix C.

In contrast, reactions in the references are different from the present invention. The process of *Barron* comprises the steps of (1) obtaining an aminosilane, (2) reacting a polyol compound with a polyisocyanate compound to produce a urethane prepolymer, and (3) reacting the aminosilane with the urethane prepolymer. (Col. 2, lines 59-66 and Col. lines 37-50.) The final product contains components of higher molecule weights and has hydrolysable silyl group, but it cannot possibly have a terminal OH group. See Appendix E.

Further, the process of *Zwiener* comprises the steps of (1) reacting a polyol compound with a polyisocyanate compound to produce a urethane prepolymer, (2) obtaining an aminosilane, and (3) reacting the amino silane with the urethane prepolymer (Examples 5 and 8).

The final product contains components of higher molecule weights and has hydrolysable silyl group, but cannot possibly have a terminal OH group. See Appendix F.

In summary, none of the cited references disclose, teach, or suggest the product of claim 46.

b. Claim 47

Since independent claim 46 is allowable, Applicants respectfully submit that claim 47 is allowable for at least the reason that it depends from an allowable claim. *In re Fine*, 837 F.2d 1071, 5 U.S.P.Q. 2d 1596, 1598 (Fed. Cir. 1988). Therefore, Applicants respectfully request that the rejection of claim 47 be withdrawn.

3. Rejection of Claim 50 under 35 U.S.C. §103

Claim 50 has been rejected under §103(a) as allegedly obvious over *Krafcik* (U.S. 5,614,604). Applicants respectfully submit that the claim amendments made herein overcome the objection. It is well established at law that, for a proper rejection of a claim under 35 U.S.C. §103 as being obvious based upon a combination of references, the cited combination of references must disclose, teach, or suggest, either implicitly, all elements/features/steps of the claim at issue. *See, e.g., In re Dow Chemical*, 5 U.S.P.Q.2d 1529, 1531 (Fed. Cir. 1988); *In re Keller*, 208 U.S.P.Q.2d 871, 881 (C.C.P.A. 1981).

1) *Krafcik* Produces a Side Reaction

In a reaction process like *Krafcik*, when a polyisocyanate is reacted with a polyol, a prepolymer is produced. If this reaction progresses ideally, the NCO terminated prepolymer (basic unit) is obtained with reaction of two polyisocyanate molecules and one polyol molecule. However, as shown by experimental data in Appendix A-1, a dimer (double unit) and trimer (triple unit) which has twice or three times of molecular weight are also produced by side

reactions, due to both polyisocyanate and polyol being multi-functional compounds.

Accordingly, the final prepolymer product contains a large amount of high weight molecule.

This is evidenced by the experimental data presented in Appendices A and B, introduced above. In Table 3-18 of Appendix A/B, chart (a) shows the prepolymer (15% isocyanate group content) which is obtained by 2:1 reaction with NCO group of polyisocyanate and OH group of polyol. Even in such prepolymer prepared at the theoretical reacting ratio, peak (7) of “pre dimer (double unit)” is observed. Further, as the isocyanate group content in the prepolymer is decreased from 15% to 6% the content of “pre dimer (double unit)” (7) and “pre trimer (triple unit)” (8) is increased. In any case where isocyanate group content is 15%, 9% or 6%, the dimer and/or trimer exist in the prepolymer product.

In other words, the side reaction, that is, production of dimerization and trimerization, *inevitably* occurs in producing a prepolymer by reacting a polyisocyanate with a polyol. Accordingly, an actual pre-polymer composition has higher molecule weight and higher viscosity than those of the ideal basic unit. Also, the final product or resin compositions, which use the prepared prepolymer as raw materials, have high viscosity. As is apparent from the above, the occurrence of side reactions is already known, and is supported by Appendix A/B.

Because a prepolymer is produced a side reaction such as dimerization and trimerization of urethane prepolymer inevitably occurs, so that a product produced by *Krafcik* has high molecule weight and high viscosity. In contrast, the process of claim 50 dispenses with the step of producing a prepolymer, and a product of claim 50 has low viscosity. Thus, the process of claim 50 differs in a non-obvious way from the processes disclosed in *Krafcik*. Accordingly, a *prima facie* case establishing an obviousness rejection under *Ex parte Rubin* has not been made.

For at least these reasons, claim 50 is not obvious over *Krafcik*, and the rejection should be withdrawn.

2) *Krafcik* Produces a Product without a Terminal OH Group

As argued in the last Office Action, according to the process disclosed in *Krafcik*, the terminal of the polymer of *Krafcik* is hydrolysable silyl group, or an alkyl group connected with a urethane linkage, and the hydrolyzed silyl group is hydrolyzed with water. In contrast, the terminal of the product of claim 50 is a hydrolysable silyl group and an OH group. (See Response filed Aug. 3, 2005, pp. 8-9). Thus, the product of *Krafcik* is different from the product of claim 50.

The outstanding Office Action stated that “applicants’ arguments are largely based upon the position that the instant products have an OH group in the terminal; however, applicants’ claims are not so limited. There is no requirement that such a terminal OH group exists or be produced.” (Office Action, p. 4.) Applicants provide herein a logical rationale supporting Applicants’ position.

Considering the process of the reaction, it is clear to one of ordinary skill in the art that the product by the process of claim 50 necessarily contains hydrolysable silyl group terminated polymer, and may in some circumstances contain OH group terminated polymer. However, Applicants do not limit the product of claim 50 to the products having a terminal OH group. Applicants’ reference to the “OH group” in the last response was made to emphasize the difference from *Krafcik*. The final product of claim 50 is hydrolysable silyl group terminated polymer, and if all ends of polyol do not react, the product may have a terminal OH group. This is simply an issue of possibility or probability.

In contrast, reactions in the references are different from the present invention. The process of *Krafcik* comprises the steps of (1) producing a urethane prepolymer, (2) reacting an

NCO group of the urethane prepolymer with an alcohol and an aminosilane, and (3) charging water (Col. 5, line 31 to Col. 6, line 9). The final product has no terminal OH group. The final product contains components of higher molecule weights and has hydrolysable silyl group, but it cannot possibly have a terminal OH group. See Appendix G. In summary, *Krafcik* does not disclose, teach, or suggest the product of claim 50.

3) Amended Claim 50 Produces a Moisture Curable Product

Applicants argued in the last response that the product of claim 50 differs from the product of *Krafcik* in that *Krafcik* is directed to a polyurethane dispersion, while the product of claim 50 is directed to a moisture curable product. The outstanding Office Action indicated that Applicants' argument "does not distinguish the claim from the prior art. Nothing in applicants' claim requires the product to be moisture curable...The claim is open to virtually any step or modification." (Office Action, p. 5, section 8.)


Claim 50 has been amended to recite "A process for the preparation of *moisture curable* urethane resins..." Therefore, even if Applicants' claimed process was analogous to changing a sequence of steps, Applicants' claimed process produces unexpected results. Therefore, a *prima facie* case establishing an obviousness rejection under *Ex parte Rubin* has not been made. For at least these reasons, claim 50 is not obvious over *Krafcik*, and the rejection should be withdrawn.

CONCLUSION

Applicants respectfully request that all outstanding objections and rejections be withdrawn and that this application and presently pending claims 46-50 be allowed to issue. If the Examiner has any questions or comments regarding Applicants' response, the Examiner is encouraged to telephone Applicants' undersigned counsel.

Respectfully submitted,

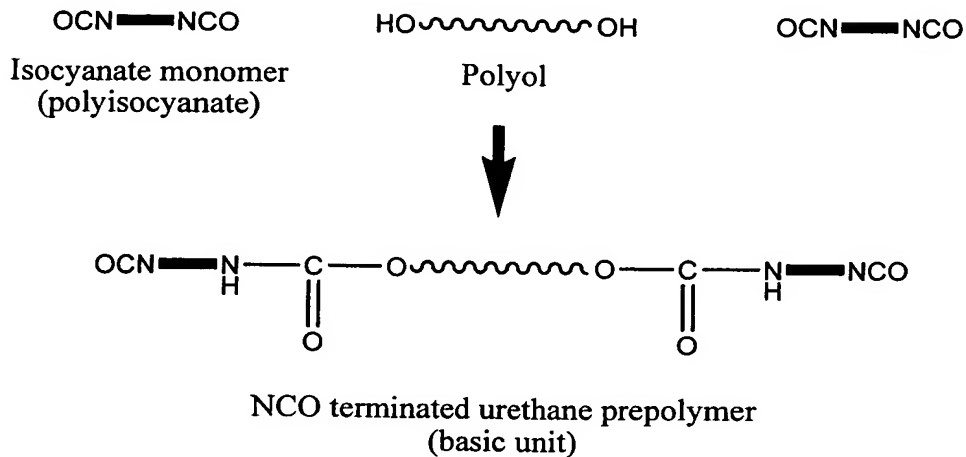
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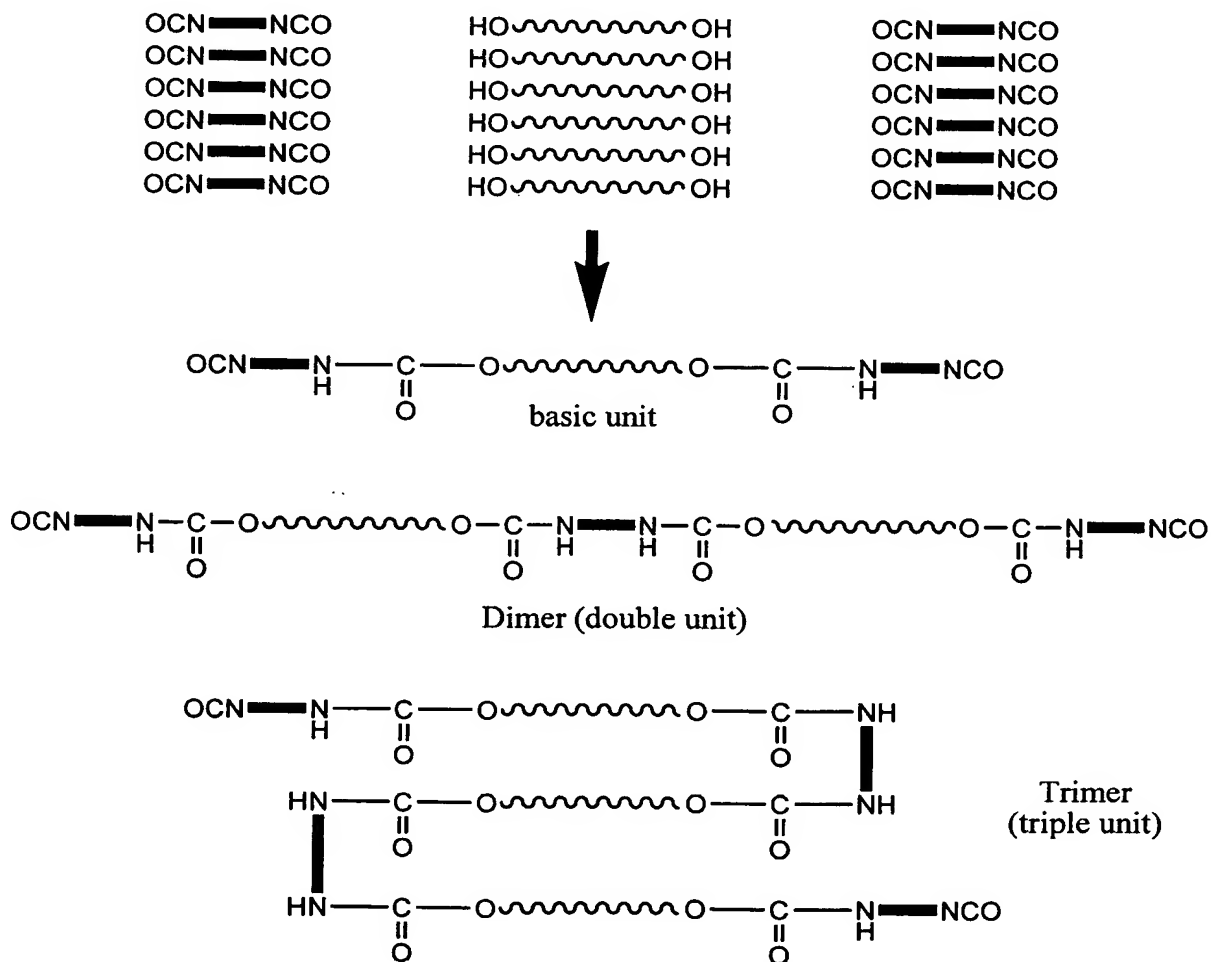
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Appendix A-1

Ideal Reaction [NCO : OH = 2 : 1]



Side Reaction (Actual Reaction) [NCO : OH = 2 : 1]



大阪では、天神祭前後が最も暑いというのが定説。今年もまた冷夏かと思われた夏
の入りも、土用を過ぎた頃から連日の猛暑、天神祭も本来の暑さのなかでの「コンチ
キチヤ」と、やっと夏らしきひなで高校野球のシーズン到来。

本号は、粘着および絶縁加工品について特集しました。粘着剤および絶縁加工品のメーカーのペタランの方に意見を頼みました。ちょっと話題の取組性はきいて、これ一冊でユーザーの方々に粘着製品の開発を理解していただけたらと思います。

この特集号が発行されるころには、秋風が吹いているかも知れませんが、でも秋気はホットに上昇してほしいものです。(1995. 7. 30) (三刀基賢)



読者のための雑誌

「接着力の技術」をより身近なものにして頂くために、ご寄
 稿やご意見を是非お寄せ下さい。

次号は1月に特集「土木工事と接点」を予定

致しております。ご期待下さい。

接 着 の 技 術 誌

平成7年9月末日発行

[illegible]

技術の支援

特集：ウレタン系接着剤

Vol. 15 No. 3 1996



江蘇

日本接着學會

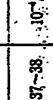




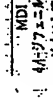




名 称	化 学 结 构	分 子 量	粘 度	玻璃(软化)点	密度g/cm ³
TDI トリレンジイソシアネート		174	3	115-135 (T-80)	10 ³ /25°C (T-80)
MDI 4,4'-メチレンジイソシアネート		250	固体	37-38	10 ³ /25°C
MDI メチルトリレンジイソシアネート		260	100-450		10 ³ /25°C以下
XDI キシレンビスイソシアネート		188	4		BP 151°C/0.8 mmHg
NDI ナフthalenビスイソシアネート		210	固体	128-130	5/16°C
PPDI フェニレンビスイソシアネート		160	固体 (134at100°C)	74-75	BP 250°C/1 mmHg
TDI トリレンジイソシアネート		244			
NDI ナフthalenビスイソシアネート		108	26	-67	10 ³ /25°C
MDI 4,4'-メチレンジイソシアネート		252	29	10-15	7×10 ³ /25°C
PPDI フェニレンビスイソシアネート		223	15	-60	7×10 ³ /20°C
LDI リジン		212			
IPC イソプロピルペンタメチレンビスイソシアネート		254	固体	48-65	
水添XDI 水添キシレンビスイソシアネート		184	6	-50	BP 110°C/0.7 mmHg
水添PPDI 水添フェニレンビスイソシアネート		165	固体 (134at165°C)	58-62	BP 250°C/1 mmHg
TDI トリレンジイソシアネート		204			

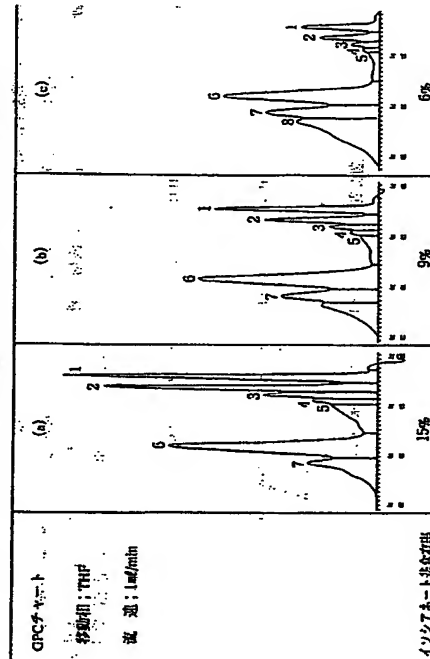
表3.16 プレポリマーの構造と硬さ

[illegible]

表3.17 原料ポリオールとウレタンプレポリマーの性状について

[illegible]

表3.18. イソシリアネート結合有単のブレイマ分子型に及ぼす影響



ピークNo.	(組成)	ポリスチレン熱算分子重		
8	(凝結体)	—	5760	6000
7	(\diamond)	4310	4310	4450
6	(結晶-7)	2550	2560	2640
5	—	—	—	—
4	(4 複体)	529	541	565
3	(3 複体)	446	456	476
2	(2 複体)	361	368	383
1	(單量体)	253	259	271
数平均分子重		567	890	1402
重平均分子重		1657	2910	4549
粘度	η_{inh}^{25} [ST]	5900	13000	15800

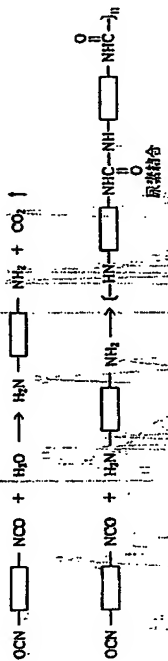
PPG (分子重3000) / ポリメリックMDI系ブレポリマ

モノマー・ジイソシアネートは芳香族と脂肪族系があり芳香族系はポリオールとの反応が強く屈折率も大きく、硬化物は黄変しやすい。脂肪族系はポリオールとの反応は速く硬化物の屈折率も小さいが黄変は無い。

ポリオロールは官能基数と分子量が使用に当たっての重要な要素であり、官能基数が高いと溶剤耐硬化物の硬さが増加する。また、分子量が高くなるとガラス転移点も高くなる。したがって、適切な物性を得るためには、官能基数と分子量を適切に調整することが重要である。

ブレポリマーの構造と種々の間隔を表3, 16に、原料ポリオールとウレタンブレポリマーの性状についてを表3, 17に、ゲルパーミエーションクロマトグラフ (GPC) からみたウレタンブレポリマーとはどのようなものなのかを表3, 18に示す。

空气中あるいは被着液中の水分と反応して硬化する一液型気硬化型ウレタン系接着剤の硬化機構を図3.20に示す。



例3.20 ウレタシン・ブレポリマーと次の反応

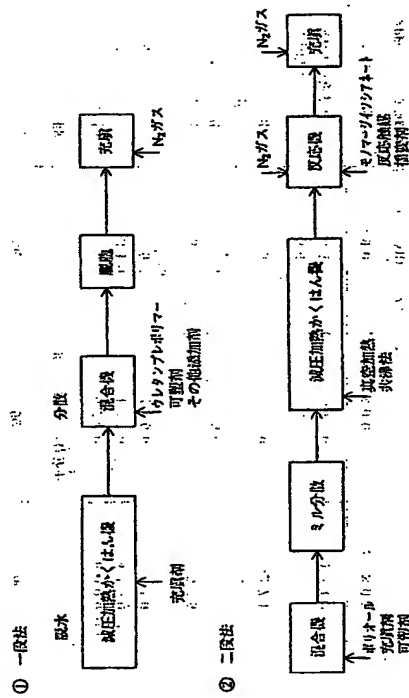


図3.21 一液湿気硬化型ウレタン系接着剤製造工程

3.3.3 一液型硬化型ウレタン系接着剤の製造方法

一般選用法として、ウレタン系接着剤の製造に際しては、重要なことは水分の除去である。特に使用する充填剤に吸着している水分は接着剤の貯蔵安定性を低下させるので、乾燥工程で安全に除去しなければならない。乾燥方法としては加熱減圧脱水処理した充填剤と、シリコン系、シリコン系等で潤滑し仕上げる2段階と、予めポリサルファー充填剤粉を混合加熱減圧脱水後、モノマー化してゆくと、1般法がある。一般的に乾燥工程を図3.21に示

3.3.4 : 一液温気硬化型ウレタン系接着剤の問題点

一 液阻硬化のため充填剤の乾燥はいうまでもなく水分と反応して炭酸ガスの発生を伴い用途により外観を損なう事や接着力の安定化に悪影響を及ぼす

す。このために発泡抑制対策が必要となる。通常その対策としては脱泡剤（モレキュラージュ・ケイ酸塩・合成ケイ酸塩・無水石膏等）脱泡ガス吸収剤として微粒子生石灰等が使用される。その他ベースプレポリマーのポリオール組成・イソシアネート含有率の影響もみのがせない。又製法システムによる対策も発表されている。図3.22は応用可能な

A. 【異種反応系】

$$\text{HOOC-NH-} \left[\text{O-CH}_2\text{-CH}_2\text{-O} \right]_n \text{-NCO} \xrightarrow{\text{ケチミンまたは}} \text{NH} \left[\text{O-CH}_2\text{-CH}_2\text{-O} \right]_n \text{NH} + \text{H}_2\text{O (水分)}$$

すなわち、異種反応系では、ケチミンまたは水と反応して、水は除去され、ケチミンが生成する。

B. 【ケチミン（アミン）系】

$$\text{HOOC-NH-} \left[\text{O-CH}_2\text{-CH}_2\text{-O} \right]_n \text{-NCO} + \text{H}_2\text{N-} \left[\text{O-CH}_2\text{-CH}_2\text{-O} \right]_m \text{-NH}_2 \rightarrow \text{NH} \left[\text{O-CH}_2\text{-CH}_2\text{-O} \right]_{n+m} \text{NH} + \text{H}_2\text{O}$$

すなわち、ケチミン系では、ケチミンと反応して、水は除去され、ケチミンが生成する。

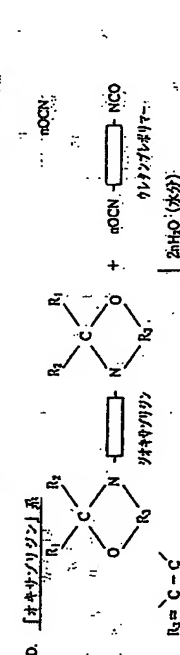
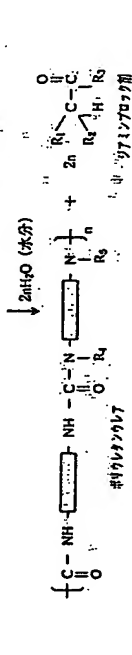
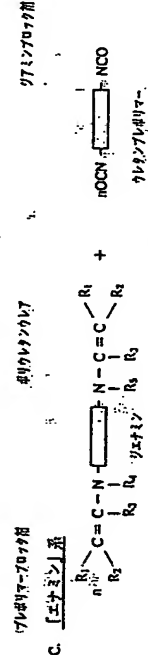
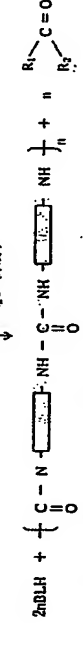
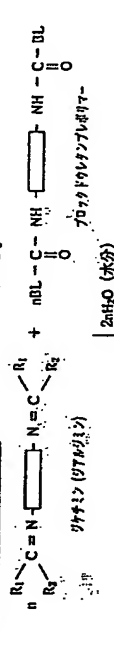
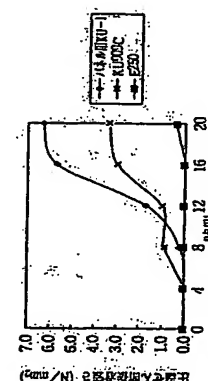


図3.22 建築用接着剤，シーラントに適用可能な湿気硬化型ポリウレタン系

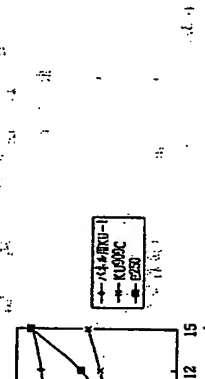
☐ : ウレタンブレポリマー骨格 ☐ : ジアミン骨格 BL : NCO プロック遊離

[illegible]

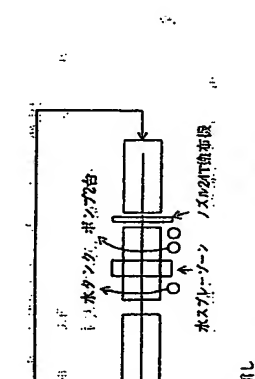
図3.24 5℃に於ける圧縮せん断接着強さ(米マツ)



100-44388-135-2



(六二) 五陽卦



钢板
0-674-129

五

 50 g/m^2

パネル型骨盤型

水ズグ・ゾー・ノ 1/1024 板本

100

板
50. m/m
50. m²

バネル製造概要

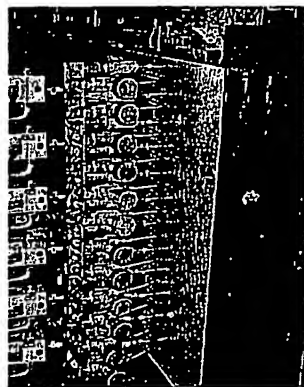


図3.27 自動ガンによるビード塗布

KU90S/Wの接着試験のデータは、表3.20に本報
構造用接着剤パネルバンド KU-1とエポキシ系接着
剤バンド E250との温度別立上り接着強度の比較を
図3.23~3.25に示す。

又自動塗布方式での KU810F/PSのパネル製造
概要図を図3.26~3.28に示す。

おわりに

一筆型硬化型ウレタン系接着剤は、無溶
剤型の界面や他の樹脂との混合化により新しい機能
を与える事により今後更に応用範囲が拡大すると思
われる。

文献

1) Polyurethanes' Chemistry and Technology Part

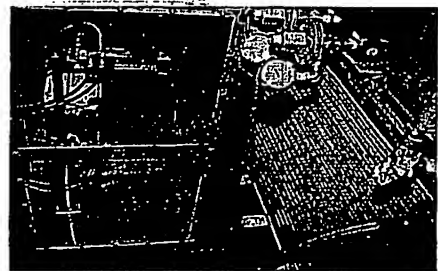


図3.28 接着剤への水スプレーゾーン

I. P. 226, p. 65 以下

2) Janusz Korakiewicz, Adhesion (GBR) 13, 114-141 (1989).

3) "ポリウレタン樹脂ハンドブック", 岩田政治編 (日刊工業新聞社).

4) 永田, (三洋化成工業株式会社), 建築技術研究会セミナ
ー 昭和63年9月12日 (東京).

5) Janusz Korakiewicz, Adhesion (GBR) 13, 114-141 (1989).



3.4 反応型二液ウレタン接着剤

堀 照 夫

はじめに

ポリウレタン (PU) 接着剤は約50年前にバイエル社が Desmodur/Desmocoll として上市して以来、イソシアネート化合物の多岐にわたる反応性、樹脂物性の多様性をはじめ優れた諸特性により年々その用途を広がってきた。今では、プラスチック・皮革・ゴム・繊維・木材・金属等に広く使用されており、その生産量は約3.3万tと書かれている。PU 接着剤がこのような多岐にわたる基材に幅広く用いられているのは、イソシアネート基が高い反応性と接着性を有し、各種活性基化合物の特性を活かした幅広い接着剤設計、ウレタン結合や環状構造の持つ高い凝集力と基材表面への物理的結合性による接着力の向上、耐水・耐薬品性などの化学的性質に優れている点に加え、常温に近い温度でも硬化が可能となるによる。

反応型二液 PU 接着剤は、一液型と比べると、配合の時間・配合液のポットライフの問題はあるものの、接着強度性の点からは優位であり、アプリケーション類の充實もあり PU 接着剤の主役を占めている。中でもその最大の用途は、食品包装用アミネット分野であり、全 PU 接着剤使用量の約1/3を占めている。

3.4.1 反応型二液 PU 接着剤の基本組成
PU 接着剤を言えばもちろん分子中にイソシアネート (NCO) 基と水酸 (OH) 基がからみあっている。NCO 基と水酸 (OH) 基を有する接着剤と定義されるが、広義には NCO 化合物より修飾されるべき接着剤を言うこともある。反応型二液 PU 接着剤では、アミン末端のポリウレタン樹脂をエポキシ基と持つ化合物で硬化するダイブ (アルカール溶剤

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系としてプリントラミ等の分野で実用化されている。ヤカルボン酸基含有ポリウレタン樹脂をカルボジイミド化合物やオキサゾリン化合物¹⁾等で硬化することも検討されているが、基本的には分子中に複数の活性水酸基 (通常は OH 基) を有する化合物一主剤と、NCO 基を有する化合物一硬化剤を、無溶剤のまま、あるいは溶剤に溶解して、あるいは水性化して用いる。さらに、用途に応じ無溶剤剤や安定剤・触媒・接着性付与剤等添加剤を用いることも多い。

3.4.1.1 活性水酸化合物
活性水酸化合物にはアミンやカルボン酸・活性メチレン基をもつものもあるが、通常 PU 接着剤に使用される原料の大部分は OH 系化合物である。表3.21に代表的な原料をあげる。反応型二液 PU 接着剤では、これらの化合物は主剤としてそのまま用いる場合とイソシアネートで活性し、分子量や凝集力を調整して用いる場合とがある (図3.29)。

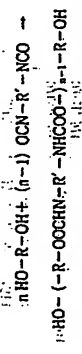


図3.29 ジョールのジイソシアネートによる変性

3.4.1.2 イソシアネート原料

PU 接着剤の製造によく用いられるジイソシアネート類を表3.22にあげる。反応型二液 PU 接着剤の硬化剤として用いられる場合には、これらジイソシアネート類は、アダグド体・ビュレト体・トリマー・ポリマー等に変性し、抽出される。抽出後は樹脂残留等の手段で非反応のジイソシアネートモノマーを除去して使用される。変性の目的は、溶剤の低下・官能基数や NCO 含有量、粘度等の調整にある。表3.23に各硬化剤の構造をあげるが、ポリメリック MDI や Desmodur IP 等の蒸気圧が低い

Appendix B

Translation

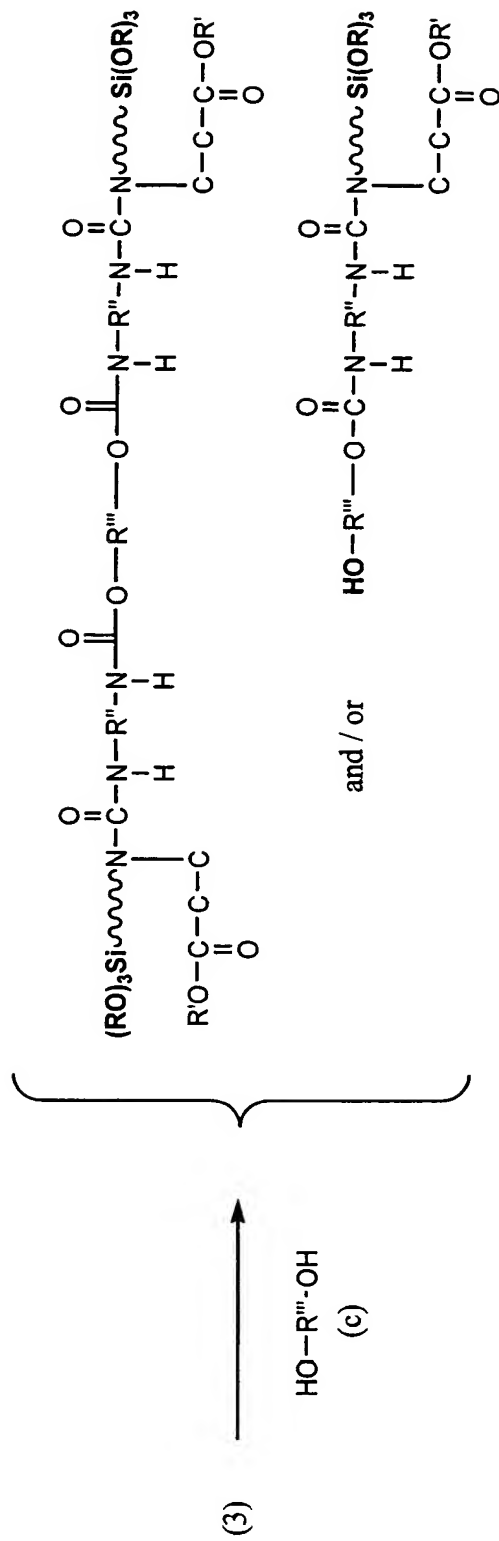
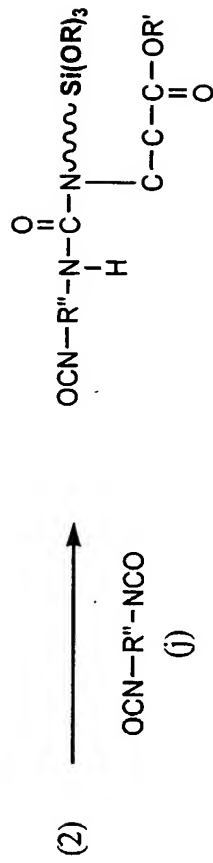
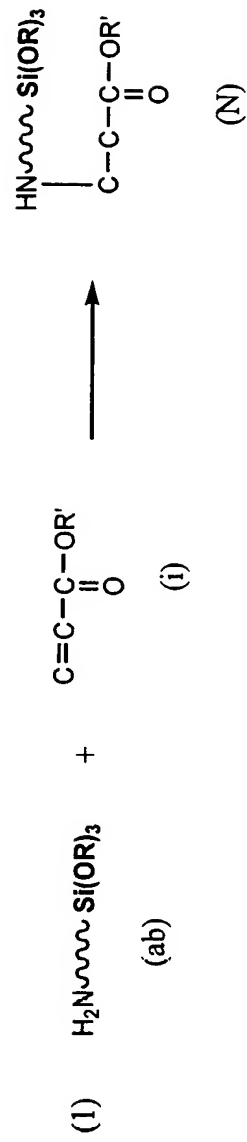
Table 3.18 Effect of Isocyanate Group Content On Molecular Weight Of Prepolymer

GPC chart Mobile phase: THF Flow rate: 1 ml/min	(a)	(b)	(c)
Isocyanate group content	15%	9%	6%
Peak No. (composition)	Molecular weight converted with polystyrene		
8 pre trimer	—	5760	6000
7 pre dimer	4310	4310	4450
6 Basic unit	2550	2560	2640
5	—	—	—
4 iso tetramer	529	541	565
3 iso trimer	446	456	476
2 iso dimer	361	368	383
1 iso monomer	253	259	271
Number average molecular weight	567	690	1402
Weight average molecular weight	1657	2910	4549
Viscosity mPa·s [25°C]	5900	13000	15800

* Polymeric MDI prepolymer

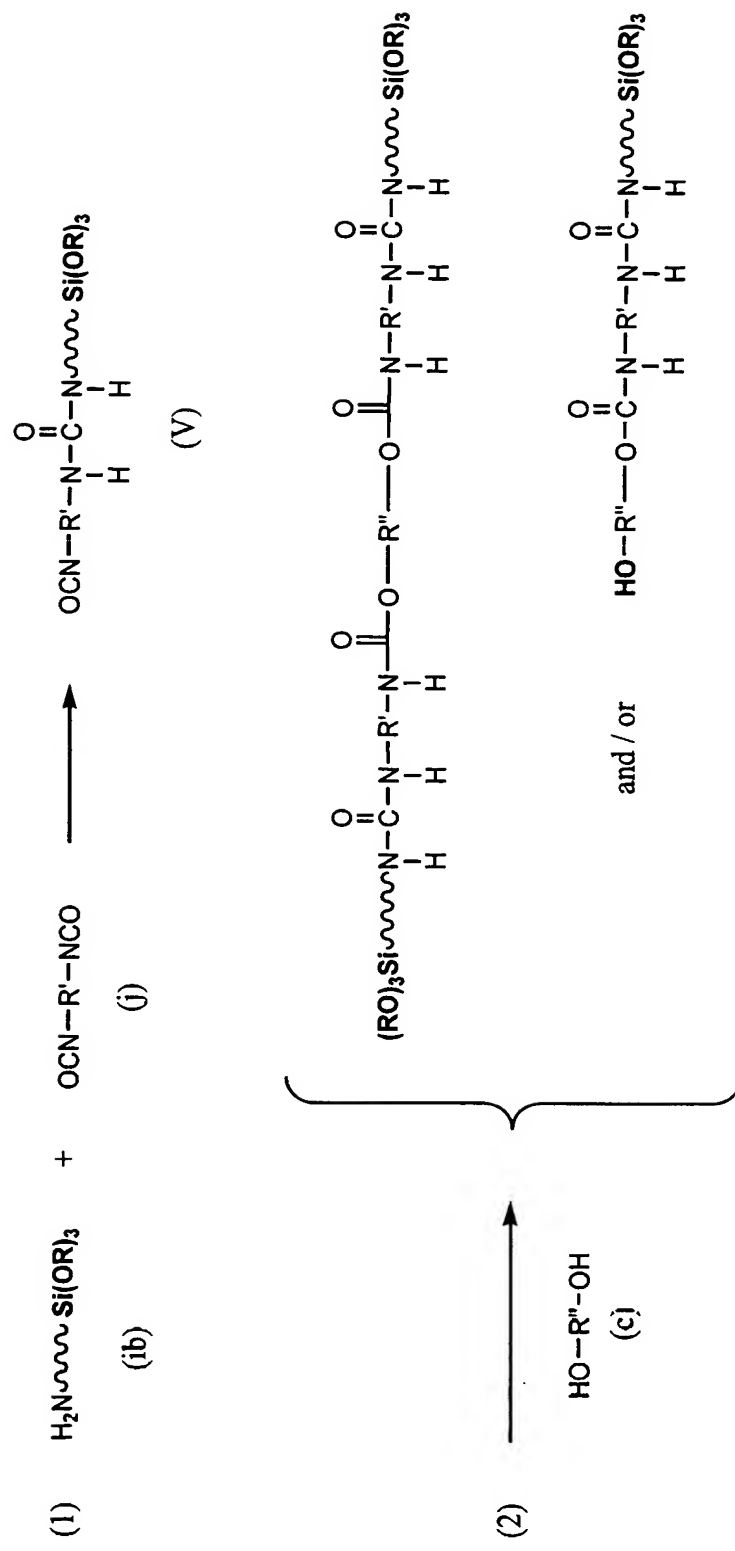
Appendix C

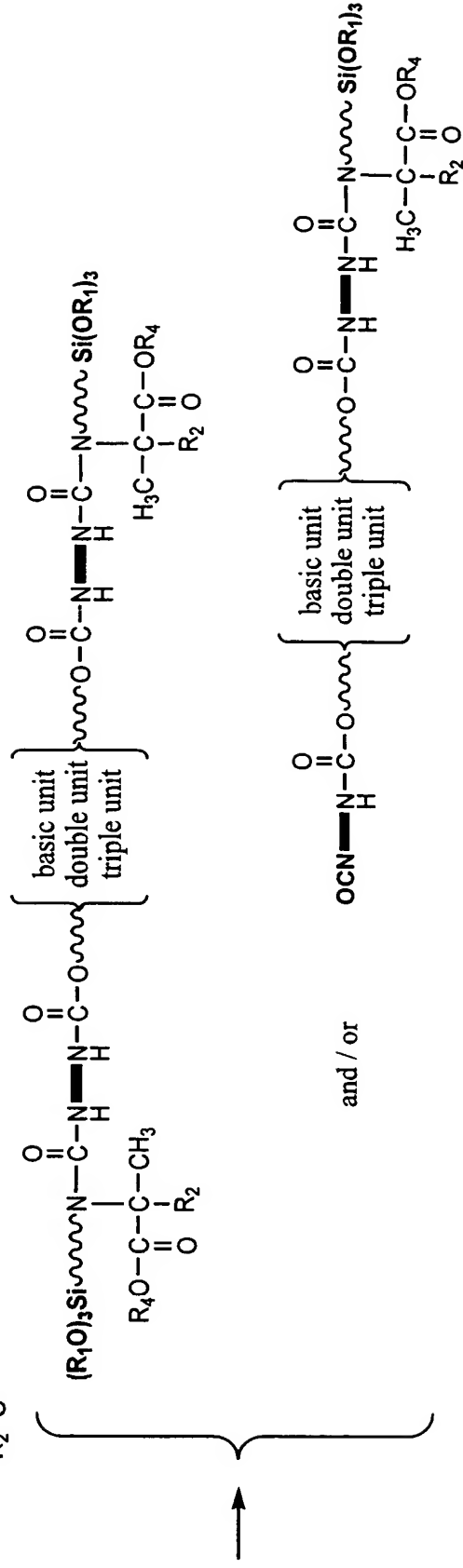
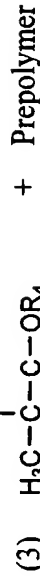
Claim 46



Appendix D

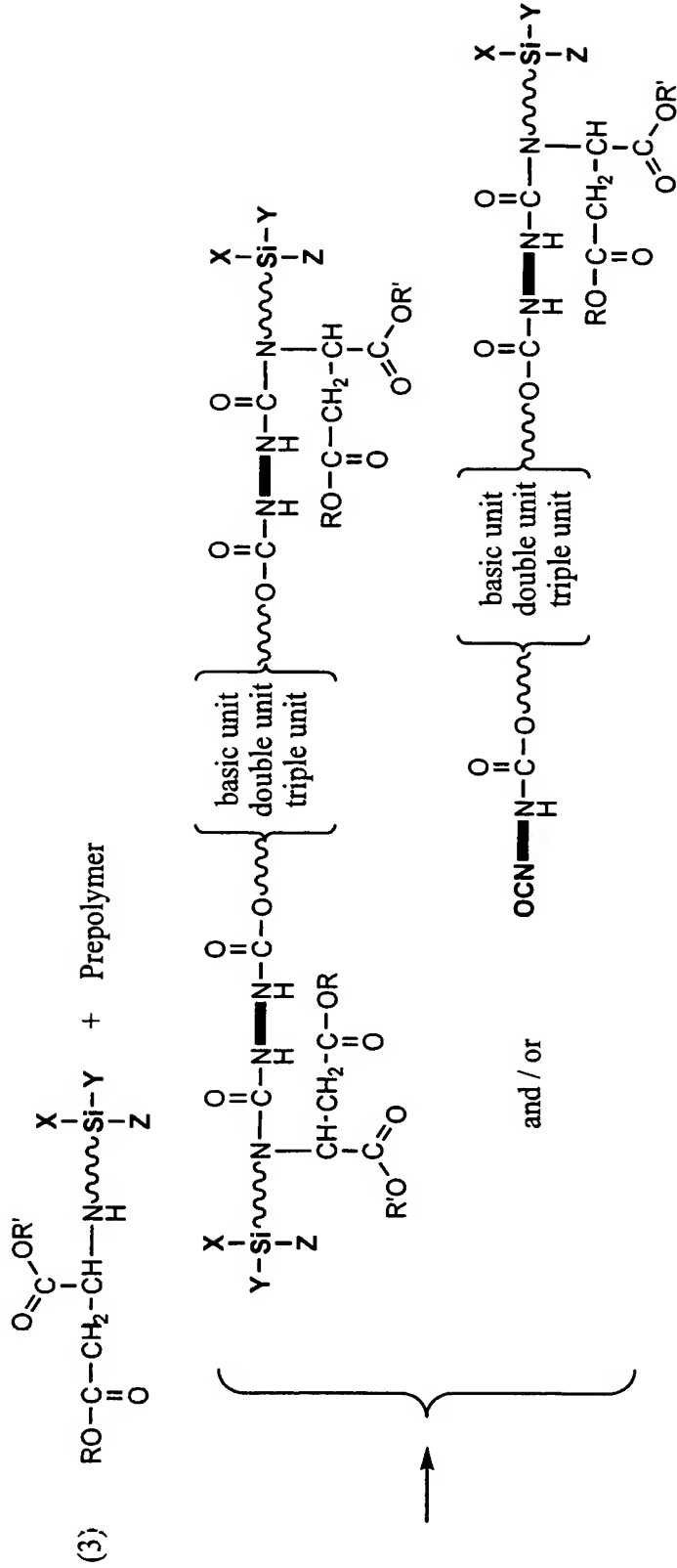
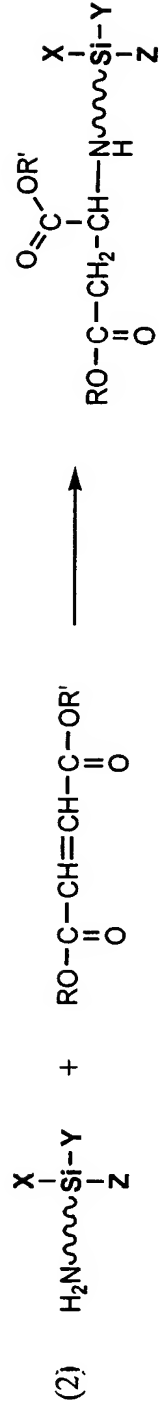
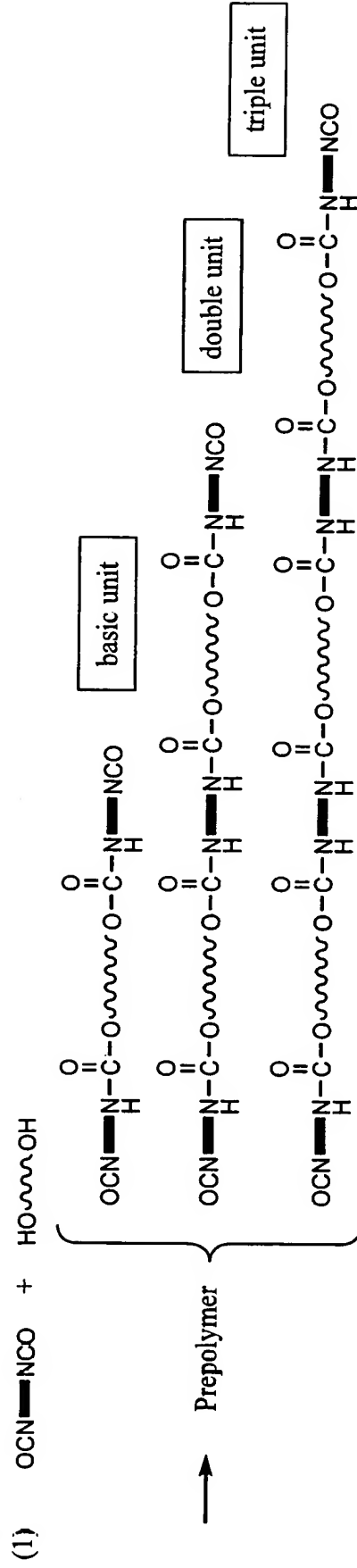
Claim 50



Barron

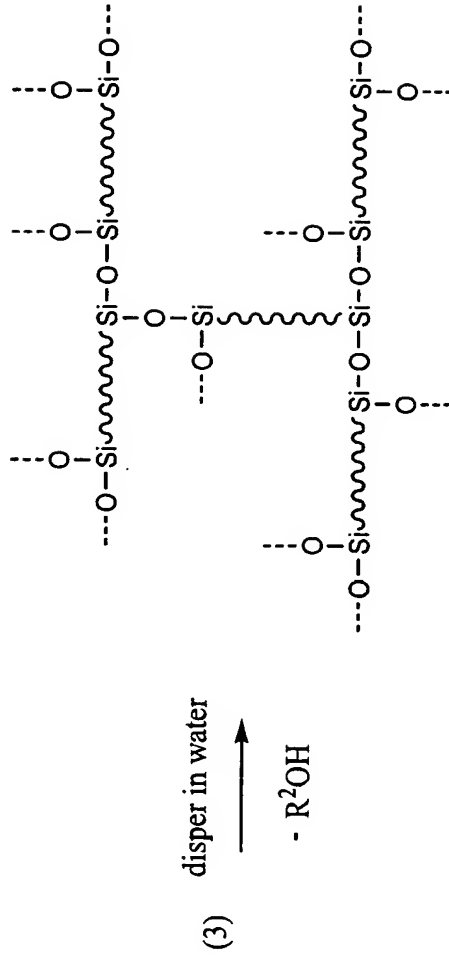
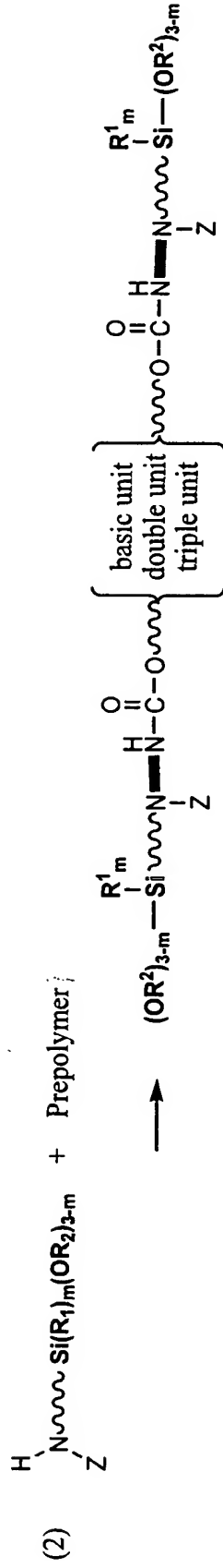
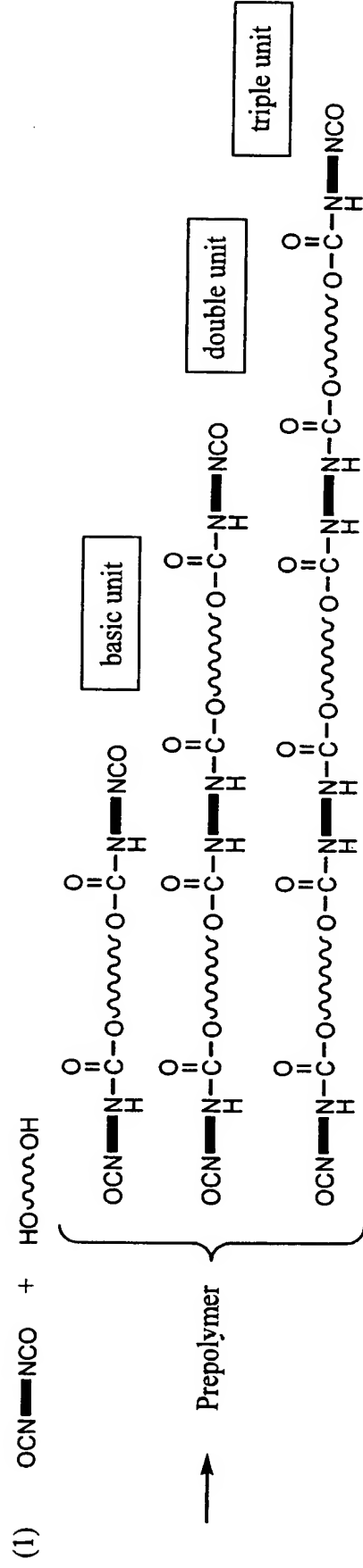
Appendix F

Zweiner



Krafcik

Appendix G



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